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X. H. Chen,¹ Y. T. Chen,¹ F.-F. Ren,^{1,2} S. L. Gu,¹ H. H. Tan,³ C. Jagadish,³ and J. D. Ye^{1,2,a)}

AFFILIATIONS

¹School of Electronic Science and Engineering, Nanjing University, Nanjing 210023, China

²Research Institute of Shenzhen, Nanjing University, Shenzhen 518000, China

³Department of Electronic Materials Engineering, Research School of Physics, The Australian National University, Canberra, ACT 2601, Australia

^{a)} Author to whom correspondence should be addressed: yejd@nju.edu.cn

ABSTRACT

Understanding the electronic structures at the interfaces of wide bandgap oxide heterostructures is crucial for the rational design of oxide-based optoelectronic devices with novel functionality and improved performance. In this work, the electronic band diagram at a ZnO/ α -Ga₂O₃ n-n isotype heterojunction is investigated by depth-profile x-ray photoemission spectroscopy (XPS). The directly measured valence-band offset is -0.61 ± 0.1 eV and a type-I (straddling gap) band alignment is formed at the ZnO/ α -Ga₂O₃ heterointerface. As probed by the depth profile of core-levels and VB-XPS, the formation of an interfacial layer is observed due to Ga and Zn interdiffusion, where charged interfacial states result in the downward and upward band-bending at the ZnO and α -Ga₂O₃ sides, respectively. The influence of band bending and band discontinuity at the interface is confirmed by the rectifying characteristics in the Au/ α -Ga₂O₃/ZnO heterojunction with electron accumulation at its interface. Taking the thermionic-field emission and band-to-band tunneling mechanisms into account, the simulated transport properties agrees well with the reported *I*-*V* characteristics of Au/ α -Ga₂O₃/ZnO avalanche photodiode, a further validation of the deduced band alignment of the heterostructure.

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Gallium oxide (Ga₂O₃) is an emerging ultrawide bandgap semiconductor with a bandgap of 4.7–5.1 eV and a high breakdown field of 8 MV cm⁻¹;^{1–3} these properties are promising for the development of high performance solar blind photodetectors and power devices.^{4–6} Despite the absence of p-type Ga₂O₃, control of band discontinuities in n-n isotype heterostructures has been proven as an alternative strategy to allow the independent manipulation of carrier injection, carrier confinement, or even ionization thresholds in Ga₂O₃-based devices.^{7–9} Among the reported polymorphs, monoclinic Ga₂O₃ (β -Ga₂O₃) is the most investigated. Band alignment of β -Ga₂O₃ with other semiconductors such as Si,¹⁰ ZnO,¹¹ GaN,¹² SiC,¹³ and AlN¹⁴ has been investigated by x-ray photoemission spectroscopy (XPS).

The metastable corundumlike α -Ga₂O₃ is attracting more attention owing to its larger bandgap (5.1 eV), higher breakdown field, and larger Baliga's figure of merit.^{15,16} To date, α -Ga₂O₃ epilayers with edge dislocation densities less than 5×10^6 cm⁻² have been achieved heteroepitaxially on sapphire through an epitaxial lateral overgrowth technique,¹⁷ and consequently, α -Ga₂O₃ based Schottky rectifiers, MOSFET power devices, and solar-blind photodetectors have been

demonstrated.^{2,3,16} To fully realize their promising potential in practical applications, a comprehensive understanding of the electronic band structures of α -Ga₂O₃ heterojunctions is crucial for the rational design of devices with multiple functionality and improved performance. Although some theoretical calculations on the electronic band structures of α -Ga₂O₃ have been conducted, the band alignment of α -Ga₂O₃ with other semiconductors has not yet been experimentally investigated.^{18,19}

X-ray photoemission spectroscopy is a powerful and commonly used technique to determine the band alignment at the interface of semiconductor heterostructures, wherein, most studies are usually focused on the determination of the valence band offset (VBO) and conduction band offset (CBO) without considering the band bending effect.^{10–14} In fact, because surfaces and interfaces are invariably imperfect, lattice defects, dangling bonds, and interdiffusion across the interface are often present.²⁰ In addition to band alignment, band bending induced by charged interfacial states has been demonstrated to influence, and sometimes dominate carrier transport and device performance. In our previous work, we reported solar-blind avalanche

photodetectors based on α -Ga₂O₃/ZnO isotype heterostructures.⁹ To obtain a comprehensive understanding of the carrier transport and device performance of the α -Ga₂O₃/ZnO isotype heterojunction, in this work we have investigated the electronic band diagram including band offsets and band bending using XPS depth profiling. The directly measured values of the VBO and CBO are -0.61 ± 0.1 eV and $(0.62-1.12) \pm 0.1$ eV, respectively, suggesting a type-I band alignment for ZnO/ α -Ga₂O₃ heterojunction. The energy shifts of core-level and VB-XPS give direct evidence of upward and downward band bending occurring at the α -Ga₂O₃ and ZnO sides, respectively. The results are attributed to charged interfacial states formed by Ga/Zn interdiffusion. In terms of the determined band alignment and band bending, the simulated transport properties are in good agreement with the experimental current-voltage (*I*-*V*) characteristics. The dominated transport mechanisms are also discussed.

The 180 nm-thick α -Ga₂O₃ epitaxial layers were grown on the *a*-plane ZnO (11 $\bar{2}$ 0) substrate by the laser molecular beam epitaxy technique, as shown by the X-ray diffraction pattern in Fig. S1 of the [supplementary material](#). The details of the growth conditions and fabrication processes have been reported elsewhere.⁹ A KrF ($\lambda = 248$ nm) pulse laser with a repetition rate of 2 Hz and a laser fluence of 1.5 J cm^{-2} was used. The *a*-plane ZnO (11 $\bar{2}$ 0) substrate has a carrier concentration of $1.2 \times 10^{17} \text{ cm}^{-3}$ and a mobility of $220 \text{ cm}^2/\text{Vs}$ at room temperature.⁹ During the growth, the substrates were mounted on a SiC heater and positioned 55 mm from the high purity Ga₂O₃ (99.999%) ceramic target. The temperature of the specimen was 600°C calibrated by an infrared thermometer and the oxygen pressure was 10^{-3} mbar. The Ga₂O₃ epilayer shows a carrier concentration of about $2 \times 10^{16} \text{ cm}^{-3}$ estimated from capacitance-voltage measurements.⁹ Prior to device fabrication, indium was evaporated on the backside of ZnO followed by metallization at 100°C for 30 min to ensure the Ohmic contact. Subsequently, Au (50 nm) Schottky contacts with diameters ranging from 100 to 400 μm were formed on the Ga₂O₃ surface by consequent processes of lithography, electron beam evaporation, and lift-off. Different Schottky contacts were separated with a spacing of about 100 μm . Finally, vertical Schottky diodes based on the $n^- - n^+$ isotype heterostructure of Au/ α -Ga₂O₃/ZnO were fabricated.

Depth-profile XPS was carried out on a bare α -Ga₂O₃/ZnO sample, which is identical to the one used for the device fabrication. XPS measurements were performed using a PHI5000 Versa Probe II electron spectrometer equipped with an Al α ($h\nu = 1486.6$ eV) x-ray radiation source and the operation pressure is about 1×10^{-6} Pa. Before XPS analysis, the sample was sputtered by an Ar⁺ ion beam at an accelerated voltage of 2 kV and emission current of 7 mA for 1 min to remove any possible surface contaminants. The sputtering process was performed with a periodic sputtering time of 60 s with an Ar⁺ ion beam raster area of $2 \times 2 \text{ mm}^2$ followed by a pause of 40 s for data acquisition until reaching the underlying ZnO. XPS data were recorded at room temperature with an energy resolution of 0.2 eV and were carefully calibrated by measuring the work function and Fermi energy (E_F) of a silver sample. The acquired XPS spectra were calibrated using the C 1s peak at 286.0 eV. *I*-*V* characteristics were measured using a Keithley source meter (model 2636B) and simulation was performed with the aid of the commercial Silvaco ATLAS program.

Figure 1(a) shows the Ga 3d core-level (CL) XPS spectra in the α -Ga₂O₃ bulk and at the ZnO/ α -Ga₂O₃ interface. After subtracting the Shirley background, the experimental XPS data can be well fitted with

FIG. 1. (a) Ga 3d core level spectra of α -Ga₂O₃ in the bulk and at the interface. (b) Zn 2p₃ core level spectra of ZnO in the bulk and at the interface. (c) Valence band spectra for the α -Ga₂O₃ thin film and ZnO substrate.

a Voigt function. The Ga 3d CL spectrum in the α -Ga₂O₃ bulk could be deconvoluted into three components. The primary peak at $E_B = 22.0$ eV corresponds to the Ga-O bond of Ga₂O₃,²¹ and a weak and broad component at 25.1 eV is due to O 2s orbital states. In addition, an asymmetric satellite peak around 20.8 eV could be associated with Ga¹(3d),²² which has been reported to be stable in β -Ga₂O₃ even upon annealing, and thus, the contribution of surface contamination can be excluded.^{21,23} The difference in binding energies of the primary Ga 3d and Ga¹(3d) CLs for α -Ga₂O₃ bulk and the ZnO/ α -Ga₂O₃ interface is about 0.3 eV in both cases. Theoretical calculations showed that the line shape of the Ga 3d peak is well reproduced by the calculated results,²³ implying that the shoulder lying at lower E_B is related to the hybridization of Ga 3d and O 2s orbital states. The XPS spectra of Zn 2p_{3/2} and Ga 3d CLs at the interface are taken from the same etching level. Figure 1(b) shows that the binding energies for Zn 2p_{3/2} CLs in the bulk and at the interface are 1022.6 and 1022.9 eV, respectively. The valence band (VB) XPS spectra of the α -Ga₂O₃ bulk and ZnO substrate are shown in Fig. 1(c), where the energy is measured from the Fermi level E_F . The O 1s spectra for α -Ga₂O₃ are broader than those of ZnO and the peak centered at a binding energy of around 11 eV is ascribed to the Zn 3d subshell emission.²⁴ The valence band maxima (VBM) for α -Ga₂O₃ bulk and ZnO are 4.38 ± 0.06 eV and 3.17 ± 0.08 eV with respect to the Fermi level, respectively; these values were obtained by linear extrapolation of the leading edges to the extended baseline of the VB spectra. The band tail like decaying emission at the edge of the VB spectrum of α -Ga₂O₃ indicates the presence of a subgap density of states (DOS) above the VBM.²¹ Indeed, the O 2s CL at the interface (shown in Fig. S2) can be deconvoluted into two major components corresponding to the coexistence of Ga-O and Zn-O bonds, which gives direct evidence of the formation of an interfacial layer such as spinel ZnGa₂O₄.^{25,26} Diffusion of Zn into Ga₂O₃ to substitute for Ga would introduce acceptors with energy levels near the VBM.²⁷⁻²⁹

Neglecting the band bending effect, ΔE_V at the ZnO/ α -Ga₂O₃ interface can be determined by the following relationship proposed by Kraut *et al.*:³⁰

$$\Delta E_V = \left[E_{\text{CL}}^{\text{Ga}_2\text{O}_3}(\text{b}) - E_{\text{V}}^{\text{Ga}_2\text{O}_3}(\text{b}) \right] - \left[E_{\text{CL}}^{\text{ZnO}}(\text{b}) - E_{\text{V}}^{\text{ZnO}}(\text{b}) \right] - \left[E_{\text{CL}}^{\text{Ga}_2\text{O}_3}(\text{i}) - E_{\text{CL}}^{\text{ZnO}}(\text{i}) \right], \quad (1)$$

where $[E_{\text{CL}}^{\text{Ga}_2\text{O}_3}(\text{i}) - E_{\text{CL}}^{\text{ZnO}}(\text{i})]$ is the energy difference between Ga 3d and Zn 2p_{3/2} CLs at the interface. $[E_{\text{CL}}^{\text{ZnO}}(\text{b}) - E_{\text{V}}^{\text{ZnO}}(\text{b})]$ and $[E_{\text{CL}}^{\text{Ga}_2\text{O}_3}(\text{b}) - E_{\text{V}}^{\text{Ga}_2\text{O}_3}(\text{b})]$ are the VBM energies with reference to the CLs in the ZnO and Ga₂O₃, respectively. Table I summarizes the CLs and VBM values for α -Ga₂O₃, ZnO, and their interface. Therefore, the VBO was directly determined to be $\Delta E_{\text{v}} = -0.61 \pm 0.1$ eV with the negative value meaning that the VBM of α -Ga₂O₃ is lower than that of ZnO. In addition, among the Ga 3d, Zn 2p_{3/2}, and O 2s CLs, any two CLs together with VBMs can be used to determine the VBO, which result in similar values of -0.61 to -0.81 eV, as shown in Fig. S2 and summarized in Table S1 of the supplementary material. In comparison, the chemical shifts of O 2s CL at the interface exhibit a larger value than those of Ga 3d CL, which are affected by multiple bonds with different metal cations. After fitting the O 1s peaks in the α -Ga₂O₃ bulk, interface, and ZnO bulk as shown in Fig. S2, the VBO of the α -Ga₂O₃/ZnO heterojunction is determined to be about -0.61 ± 0.1 eV, consistent with the VBO value determined by Ga 3d and Zn 2p_{3/2}. Consequently, the conduction band offset (CBO) can be calculated using $\Delta E_{\text{c}} = E_{\text{g}}^{\alpha\text{-Ga}_2\text{O}_3} - E_{\text{g}}^{\text{ZnO}} - |\Delta E_{\text{v}}|$. The bandgaps of ZnO and α -Ga₂O₃ used in this work are 3.37 eV (Refs. 31–33) and 5.1 eV,³ respectively, as determined by transmittance spectra in Fig. S3. Therefore, the conduction band level of α -Ga₂O₃ is higher than that of ZnO with the estimated CBO of 1.12 ± 0.1 eV. If the interfacial ZnGa₂O₄ with a lower direct bandgap of 4.6 eV is considered,³⁴ the CBO is reduced to be 0.62 ± 0.1 eV. The effective tailoring of the band alignment has been also reported in the Ga_xZn_{1-x}O/InGaZnO heterojunction by tuning the Ga component and the VBO and CBO values for Ga_{0.47}Zn_{0.53}O_{1.14}/IGZO are consistent with values in this work if the effect of the interfacial layer is taken into consideration.³⁵ Regardless of the formation of an interfacial layer, a type-I band alignment (straddling gap) is found for the ZnO/ α -Ga₂O₃ heterojunction. The large band discontinuity at both the VB and CB dominates carrier injection and transport properties, which is expected to produce rectifying characteristics similar to a Schottky diode or p-n junction and allow for the design of optoelectronic devices without a p-n junction.

The determination of VBO and CBO discussed above has not taken the band bending at the heterointerface into account. In the presence of an interfacial layer with charged states involved, electronic band bending could be quite different from what is expected from a simple comparison of the work functions or electron affinities at the thermodynamic equilibrium. A previous study has shown that the substrate cation peaks shift rigidly with the valence band features, so that both shifts are due to band-bending changes.³⁶ For a more precise determination, a possible interface-induced change in band bending

TABLE I. Peak position of core levels and VBM used to calculate the VBO in the ZnO/ α -Ga₂O₃ heterojunction.

Sample position	Region	Binding energy (eV)
α -Ga ₂ O ₃ bulk	Ga 3d	22.0
	VBM	4.38 ± 0.06
ZnO bulk	Zn 2p ₃	1022.6
	VBM	3.17 ± 0.08
α -Ga ₂ O ₃ /ZnO interface	Ga 3d	21.7
	Zn 2p ₃	1022.9

must be considered by monitoring the shift of CLs and VBMs in the bulk and at the interface. Depth-profile XPS spectral maps for VB, Ga 3d, O 1s, and Zn 2p₃ CLs of the ZnO/ α -Ga₂O₃ heterojunction are shown in Fig. 2. Overall, the depth distribution of Ga 3d and Zn 2p₃ CLs exhibits a larger overlap on the α -Ga₂O₃ side than the ZnO side with a total overlap width of about 30 nm. It is expected that the epitaxial process promotes the out-diffusion of Zn from the ZnO substrate into the growing α -Ga₂O₃ layer.^{26,28,37} A solid-phase reaction at the interface also facilitates the formation of a ZnGa₂O₄ interfacial layer. As expected, the peak position of the Ga 3d CL clearly shifts to lower binding energy from α -Ga₂O₃ bulk to the interface, indicating an upward band bending of $E_{\text{bb}}(\text{Ga}_2\text{O}_3) = 0.3$ eV on the α -Ga₂O₃ side, while the spatial evolution of the Zn 2p₃ CL clearly implies a downward band bending of $E_{\text{bb}}(\text{ZnO}) = 0.3$ eV on the ZnO side.³⁸ It is also clearly seen that the binding energy shift of the VBM (dominated by O 2p orbitals) and the O 1s CL from α -Ga₂O₃ to ZnO bulk side is about 1.22 ± 0.1 and $1.30 \text{ eV} \pm 0.1$ eV, respectively, which equals the sum of the band-bending induced built-in potential of 0.6 eV and the VBO of 0.61 eV at the interface.

A similar band-bending effect has been observed in the Ga_{0.47}Zn_{0.53}O_{1.14}/IGZO interface and could be understood in terms of an interfacial charge model.³⁵ Ga substitution for Zn in ZnO is well known to act as a shallow donor while Zn in Ga₂O₃ acts as a deep-level acceptor. Similar to the built-in field formed in p-n junctions, charged dipoles are formed through the charge transfer from donor-type bonds to acceptor-type bonds in the interface region. In this case, the charge equilibration process would result in the filling and/or emptying of defect levels until the highest occupied donor or lowest acceptor states are positioned at the Fermi level. Therefore, taking band-bending and band edge discontinuity into account, the schematic energy band diagram of the ZnO/ α -Ga₂O₃ heterojunction in

FIG. 2. Spectral map for VB, Ga 3d, O 1s, and Zn 2p₃ CLs of the ZnO/ α -Ga₂O₃ heterojunction.

thermal equilibrium is shown in Fig. 3, where E_c is the conduction band minimum, E_v is the valence band maximum, E_{CL} is the core level energy, and E_g is the energy bandgap. The respective upward and downward band-bending at the α -Ga₂O₃ and ZnO sides leads to a reduced barrier for electrons in the conduction band. Specifically, the CBM of ZnO even extends close to the Fermi level at the interface and electrons could accumulate in the conduction band.

To validate the calculated band diagram at the interface of ZnO/ α -Ga₂O₃ heterojunction, carrier transport and interface properties of the Au/ α -Ga₂O₃/ZnO/In heterojunction are revisited. Simulated I - V characteristics by the Silvaco ATLAS program are shown in Fig. 4(a) together with the experimental I - V feature. For simulation, an Ohmic contact and a Schottky contact were used for ZnO and α -Ga₂O₃, respectively. The electron affinity, bandgap, effective density of states in the conduction band and valence band, permittivity, and the electron mobility of α -Ga₂O₃ used in the Silvaco ATLAS simulations were set to be 3.9 eV,^{39,40} 5.1 eV, $3.72 \times 10^{18} \text{ cm}^{-3}$, $3.72 \times 10^{18} \text{ cm}^{-3}$, 10.0 ϵ_0 , and 10 cm²/V s, respectively, where ϵ_0 is the vacuum permittivity.⁴¹ The parameters and values of ZnO used in the simulation were the default ones in the Silvaco ATLAS database, in which the electron mobility and the affinity were set as 220 cm²/V s and 4.6 eV, respectively. The work function of Au (Schottky contact) was set as 5.3 eV.⁴² When taking the formation of ZnGa₂O₄ at the interface into account, the CBO at the interface is set to be 0.7 eV due to the bandgap lowering effect. The simulated I - V characteristics closely reproduce the experimental results in the forward bias region. The discrepancy in the reverse bias region is due to the limitation of instrument noise floor of 0.1 pA. When the Au/Ga₂O₃ Schottky contact is forward biased less than 10 V, the current follows an exponential relationship, which is well described by the thermionic-field emission mechanism. As shown by the schematic band diagram in the Fig. S4, this result is mainly attributed to the blocking effect of electrons injected from ZnO into α -Ga₂O₃ due to the presence of an additional barrier induced by the band bending and CBO at the ZnO/ α -Ga₂O₃ heterojunction interface. A kink in the I - V curve is noticeable at a forward bias of about 10 V, beyond which the current rises rapidly. It is a result of band-to-band tunneling process through the barrier from ZnO into α -Ga₂O₃ in the presence of charged defects induced by interdiffusion at the

FIG. 3. Schematic energy band diagrams of the ZnO/ α -Ga₂O₃ heterojunction with band-bending in thermal equilibrium.

FIG. 4. (a) Experimental and simulated I - V characteristics of the Au/ α -Ga₂O₃/ZnO avalanche photodiode. (b) The simulated band structure and electron concentration profiles at zero bias.

interface.^{43–45} The simulated electronic band structure of the ZnO/ α -Ga₂O₃ heterojunction and the depth profile of carrier concentration at zero bias are shown in Fig. 4(b). The values of band-bending and band discontinuities are very similar to those of Fig. 3, in which an upward and downward band-bending is formed at the α -Ga₂O₃ and ZnO side, respectively. Electron accumulation occurs at the ZnO side with a peak value of about 10^{18} cm^{-3} . Indeed, further polarization engineering could be implemented if the growth of α -Ga₂O₃ is carried out on ZnO (0001) polar substrates, which could change the band bending effect and enhance the concentration of a two-dimensional electron gas (2DEG) at the interface for the high mobility transistor applications.^{46,47}

In summary, the electronic band structure alignment of the ZnO/ α -Ga₂O₃ n-n isotype heterojunction has been determined experimentally by depth-profile XPS. A type-I heterojunction was deduced with a directly measured VBO of $-0.61 \pm 0.1 \text{ eV}$. Due to the interdiffusion of Ga and Zn into the opposite sides of the heterojunction, an interfacial layer is formed. The band-bending effect in ZnO and α -Ga₂O₃ was observed in the interface region, as a result of charge transfer and formation of dipoles in the interfacial layer. Simulated transport properties reproduced the experimental I - V characteristics of the Au/ α -Ga₂O₃/ZnO heterostructure very well, verifying the accuracy of the band alignment and band-bending, which allows us to better design wide-bandgap oxide heterostructures for advanced deep-ultraviolet applications.

See the [supplementary material](#) for detailed results of XRD, XPS, transmittance, and the schematic band diagram of the α -Ga₂O₃/ZnO heterostructure and the determined VBO values were summarized in Table S1.

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